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**Chemical markers for the authentication of unifloral *Salvia officinalis* L. honey**

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**Highlights**

- Chemical characterization of unifloral *Salvia officinalis* L. honey
- Determination of polyphenolics, carbohydrates and minerals
- Chemical markers for the authentication of *Salvia officinalis* honey

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1    **ABSTRACT**

2    The objective of the present study was to trace phytochemicals that characterize unifloral  
3    Common sage (*Salvia officinalis* L.) honey originating from the Croatian North Adriatic  
4    coast. The polyphenolic profiles and total phenolic contents (TPC), the compositions of  
5    minerals, sugars and sugar alcohols, and the radical scavenging activities (RSA) of 18  
6    unifloral *S. officinalis* honey samples were investigated. The quantitative data on the targeted  
7    compounds (25 phenolic compounds, 14 carbohydrates and 25 minerals) together with the  
8    TPC and RSA data served as a pool of variables for multivariate analysis, which provided  
9    useful information for the accurate authentication of unifloral sage honey and its  
10   discrimination from other unifloral types of honey. The proposed markers, together with  
11   chemometrics, could further contribute, as a powerful tool, to the quality control of Croatian  
12   unifloral *S. officinalis* honey and thus, possibly certify its commercial value.

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16    **Keywords:** Unifloral honey, *Salvia officinalis* L., Chemical markers, Polyphenolics, Sugars,  
17    Sugar alcohols, Minerals, Food analysis, Food composition

18

19     **1. Introduction**

20

21         Common sage (sometimes called Great sage or Dalmatian sage, Latin *Salvia*  
22         *officinalis* L.) is a circum-Mediterranean nectariferous botanical species common to the  
23         Eastern Adriatic and Ionian seas (Ricciardelli D'Albore and Galarini, 2000) with a habitat  
24         reaching south into northwest Greece (Karousou et al., 2000). This spontaneous perennial  
25         Mediterranean shrub (belonging to the family *Lamiaceae*), widespread in the Mediterranean  
26         part of Croatia, spontaneously grows on the hillsides of the North Croatian Littoral and  
27         Dalmatian islands as well ~~in~~on the adjacent coastal belt (800–5000 m wide) and has  
28         significant beekeeping importance (Flora Croatica Database, 2012). The North–East part of  
29         the Adriatic Littoral (North Croatian Littoral) is especially characterized by an abundance of  
30         sage-dominated botanical communities. Actually, they sometimes cover areas of several  
31         square kilometers, representing practically ~~the~~by far the most predominant plant of this poor,  
32         rocky terrain of the karst region (Šugar et al., 1983). Through the ~~centennial~~ tradition of  
33         beekeeping, this area has become well known for its famed unifloral *S. officinalis* honey that  
34         has seen widespread use in traditional medicine for the treatment of respiratory problems, as  
35         an antiseptic, *etc.* The potential health effects of this unifloral honey are usually ascribed to  
36         its phytochemical constituents, which mostly originate from *S. officinalis* nectar (Kenjerić et  
37         al., 2008).

38         The objective of the present study was to determine useful chemical markers for the  
39         authentication of unifloral *S. officinalis* honey, based on the analysis of the polyphenolic  
40         profiles, minerals, sugars and sugar alcohols in 18 honey samples originating from the  
41         North~~north~~east Adriatic region of Croatia. The phytochemical profiles of the studied  
42         honey samples were analyzed by high resolution LC/MS techniques. Quantification of major  
43         phenolic compounds was achieved using ultra-ultra-high-performance liquid chromatography

44 coupled with a diode array detector and a triple quadruple mass spectrometer (UHPLC DAD–  
45 MS/MS). In order to trace the phytochemicals that characterize sage honeys produced in the  
46 North Croatian Littoral, this work was focused on the identification of target compounds  
47 using ~~ultra~~-ultra-high-performance liquid chromatography coupled with hybrid mass  
48 spectrometry, which combined a Linear Trap Quadrupole and OrbiTrap mass analyzer  
49 (UHPLC–LTQ OrbiTrap MS). This technique has already proven itself to be reliable for the  
50 unambiguous detection of phenolic acids and their derivatives, as well as of the flavonoids  
51 aglycones and glycosides. The sugar content was determined using high-performance anion-  
52 exchange chromatography with pulsed amperometric detection (HPAEC/PAD). The  
53 characterization of ~~Common~~-common sage unifloral honey was further supported by the  
54 evaluation of the mineral composition using inductively coupled plasma-atomic emission  
55 spectroscopy (ICP-OES) and melissopalynological analysis.

56

## 57 **2. Experimental**

58

### 59 *2.1. Sage honey sampling and the authenticity of the samples*

60 Representative honey sampling was performed directly at the filling facilities of ~~the~~  
61 primary producer. After ~~sampling~~collection, samples were placed into α-glass jars sealed with  
62 ~~the~~ metal lids and kept at temperature of +4 °C to +8 °C until analyzed. In order to attain  
63 confirmation of the botanical origin of the *S. officinalis* honeys, the samples were subjected to  
64 thorough melissopalynological and sensory assessment. Melissopalynological analysis,  
65 considered as an analytical tool essential for the verification of the botanical and geographical  
66 origin of a honey, was realized according to the method described by Loveaux et al. (1978)  
67 and further elaborated by Von der Ohe et al. (2004).

68        The extent to which a honey sample corresponds to a given plant source is determined  
69        from the frequencies of the pollen and honeydew elements in it. Since sage pollen is under-  
70        represented, and the percentage of sage pollen in the sediment is lower than the percentage of  
71        the corresponding nectar in the honey (Ricciardelli D'Albore and Galarini, 2000), the  
72        melissopalynological assessment was based on the expression of the pollen representativity  
73        within pollen frequency classes: "predominant pollen" (more than 45-% of the pollen grains);  
74        "secondary pollen" (16–45 %); "important minor pollen" (3–15 %); minor pollen" (less than  
75        3 %), as well as on the presence of honeydew elements (Loveaux et al., 1978; Von der Ohe et  
76        al., 2004). Sensory assessment, as an equally important analytical mechanism for the  
77        determination of the unifloral character of a honey (Piana et al., 2004), comprehensive  
78        distinctive organoleptic features (visual, taste, odour, tactile) of the samples were determined  
79        taking into consideration the extent of their compliance with the organoleptic profile of  
80        unifloral sage honey (Lušić et al., 2007).

81

## 82        2.2. Reagents and standards

83        Acetonitrile and formic acid (both MS grade), methanol (HPLC grade), Folin—  
84        Ciocalteu reagent, sodium carbonate, hydrogen peroxide, and hydrochloric and nitric acid  
85        were purchased from Merck (Darmstadt, Germany). Trolox (6-hydroxy-2,5,7,8-  
86        tetramethylchroman-2-carboxylic acid) was purchased from Sigma Aldrich (Steinheim,  
87        Germany). 2,2-Diphenyl-1-picrylhydrazyl·(DPPH·) was purchased from Fluka AG (Buchs,  
88        Switzerland). The Strata C18—E (500 mg/3mL) SPE cartridges used for the extraction and  
89        concentration of samples were obtained from Phenomenex (ThermoFisher  
90        Scientific Torrance, CA). Ultra-pure water (Thermofisher TKA MicroPure water purification  
91        system, 0.055 µS/cm) was used to prepare the standard solutions and blanks. Syringe filters  
92        (13 mm, PTFE membrane 0.45 µm) were purchased from Supelco (Bellefonte, PA, USA).

93           *cis, trans*-Abscisic acid and polyphenolic standards were purchased from Fluka AG  
94           (Buchs, Switzerland). Sugar standards were purchased from Tokyo Chemical Industry  
95           (ZwijndrechtTCI, Europe, Belgium) and sugar alcohol standards were obtained from Sigma  
96           Sigma-Aldrich (Steinheim, Germany).

97

98           *2.3. Preparation of standard solutions*

99           A 1000 mg/L stock solution of a mixture of all phenolic standards and *cis, trans*-  
100          abscisic acid was prepared in methanol. Dilution of the stock solution with methanol yielded  
101          the working solutions of concentrations 0.025, 0.050, 0.100, 0.250, 0.500, 0.750, and 1.000  
102          mg/L. Calibration curves were obtained by plotting the peak areas of the standards against  
103          their concentrationCalibration curves were obtained by plotting the peak areas of the  
104          compounds identified relative to the peak area against the concentration of the standard  
105          solution. Calibration curves revealed good linearity, with  $R^2$  values exceeding 0.99 (peak  
106          areas vs. concentration).

107          The evaluation of the carbohydrate content of the honey samples was obtained from  
108          calibration curves of pure compounds. The calibration was performed with standard solutions  
109          of sugars and sugar alcohols dissolved in ultrapure water. Each individual standard was  
110          dissolved in ultrapure water. Stock solutions with concentrations of 1000 mg/L were prepared  
111          and working solutions in the concentration ranges were as follows: for glucose and fructose  
112          from 10.0 to 100.0 mg/L; for sucrose from 1.0 to 10.0 mg/L; for isomaltose from 0.5 to 5.0  
113          mg/L, while for all the other standards, the concentration range was from 0.1 to 1.0 mg/L.  
114          Under these chromatographic conditions, the last compound was detected after approximately  
115          25 min, and the analysis was ended at 30 min.

116 To analyze the mineral composition of honey, a multi-element plasma standard  
117 solution 4, Specpure, containing 1 g dm<sup>-3</sup> of each element was utilized for reference  
118 purposes.

119

120 *2.4. LC–MS/MS analysis*

121 *2.4.1. Preparation of sample extracts*

122 The method previously described by Gasic et al. (2014) was used for extraction and  
123 isolation of phenolics from the honey samples. Prior to UHPLC–DAD MS/MS and UHPLC–  
124 MS/MS Orbitrap analysis, the extracts were filtered through a 0.45–45- $\mu$ m PTFE membrane  
125 filter.

126

127 *2.4.2. UHPLC–MS/MS Orbitrap analysis of polyphenolic compounds*

128 Separation of the compounds of interest were was performed using a liquid  
129 chromatography system that consisted of a quaternary Accela 600 pump and an Accela  
130 Autosampler, connected to a linear ion trap–orbitrap hybrid mass spectrometer (LTQ  
131 OrbiTrap XL) with a heated-electrospray ionization probe, HESI-II (ThermoFisher  
132 Scientific, Bremen, Germany).

133 A Syncronis C18 column (100 × 2.1 mm, 1.7  $\mu$ m particle size) from Thermo Fisher  
134 Scientific was used as the analytical column for separation. The mobile phase consisted of  
135 (A) water + 0.1% formic acid (A) and acetonitrile + 0.1% formic acid (B)water + 0.1 %  
136 formic acid and (B) acetonitrile + 0.1 % formic acid. A linear gradient program at a flow rate  
137 of 0.300 mL/min was used: 0.0–1.0 min 5 % B, 1.0–9.9 min from 5 % to 95 % (B), 9.9–10  
138 min from 95 % to 5 % (B), then 5 % (B) for 3 min. The injection volume was 5  $\mu$ L (Gasic et  
139 al., 2014).

140 The mass spectrometer was operated in the negative ion mode. The HESI-source  
141 parameters were given previously (Gasic et al., 2014). Xcalibur software (version 2.1) was  
142 Xcalibur software 2.1 (Thermo Fisher, Bremen, Germany) was used for instrument control,  
143 data acquisition and data analysis. The phenolics were identified according to the  
144 corresponding spectral characteristics: mass spectra, accurate mass, characteristic  
145 fragmentation, and characteristic retention time. Full scan analysis was employed to detect  
146 the monoisotopic mass of unknown compounds, while the fragmentation pathway was  
147 obtained by MS/MS. This exact mass search method was based on high resolution MS  
148 analysis (Orbitrap), online database search (Patiny and Borel, 2013) and prediction of  
149 MS/MS fragmentation using Mass Frontier 6.0 software (Thermo Fisher Scientific).

150

#### 151 2.4.3 UHPLC-DAD MS/MS analysis of polyphenolic compounds

152 The separation, determination, and quantification of the components in the sage honey  
153 samples were performed using a Dionex Ultimate 3000 UHPLC system equipped with a  
154 diode array detector (DAD) that was connected to TSQ Quantum Access Max triple-  
155 quadrupole mass spectrometer (ThermoFisher Scientific, Basel, Switzerland). The elution  
156 was performed at 40 °C on a Syncronis C18 column. The mobile phase consisted of (A)  
157 water + 0.1 % formic acid (A) and acetonitrile (B) + 0.1 % formic acid, and (B)  
158 acetonitrile, which were applied in the following gradient elution: 5 % B in the first 2.0 min,  
159 2.0–12.0 min 5–95 % B, 12.0–12.2 min from 95 % to 5% B, and 5 % B until the 15<sup>th</sup> min.

160 The flow rate was set to 0.4 mL min<sup>-1</sup> and the detection wavelengths to 254–  
161 and 280 nm. The injection volume was 5 µL.

162 A TSQ Quantum Access Max triple-quadrupole mass spectrometer equipped with an  
163 heated electrospray ionization (HESI) source was used with the vaporizer temperature kept at  
164 200 °C, and the ion source settings as follows: spray voltage 5000 V, sheet gas (N<sub>2</sub>) pressure

165 40 AU, ion sweep gas pressure 1 AU and auxiliary gas ( $N_2$ ) pressure 8 AU, capillary  
166 temperature 300 °C, and skimmer offset 0 V\_(Natic et al., 2015). The mass spectrometry data  
167 were acquired in the negative ion mode, in the  $m/z$  range from 100 to 1000. Multiple mass  
168 spectrometric scanning modes, including full scanning (FS), and product ion scanning (PIS),  
169 were conducted for the qualitative analysis of the targeted compounds. The collision-induced  
170 fragmentation experiments were performed using argon as the collision gas, and the collision  
171 energy was varied depending on the compound (**Table S1**). The time-selected reaction  
172 monitoring (tSRM) experiments for quantitative analysis were performed using two  $MS^2$   
173 fragments for each compound that were previously defined as dominant in the PIS  
174 experiments (**Table S1**).

175 Xcalibur software 2.2 (Thermo Fisher, Bremen, Germany) Xcalibur software (version  
176 2.2) was used for instrument control. The phenolics were identified by direct comparison  
177 with commercial standards. The total amounts of each compound were evaluated by  
178 calculation of the peak areas and are expressed as  $mg\ kg^{-1}$ .

179

#### 180 2.5. Determination of TPC and RSA

181 The samples were prepared according to a previously described method (Gasic et al., 2014).  
182 Each honey sample (5 g) was mixed with ultrapure water in a 50-mL volumetric flask. The  
183 solution was then filtered through 0.45- $\mu$ -m PTFE membrane and analyzed for determination  
184 of TPC and RSA. -The amount of total phenolics was determined according to the Folin—  
185 Ciocalteu method, while the radical-scavenging activity of honey extracts was measured  
186 using the DPPH· method (Gasic et al., 2014). The TPC and RSA values are expressed as  
187 milligram gallic acid equivalents (mg GAE) per equivalents (GAE) per kilogram and  
188 micromoles of Trolox equivalents ( $\mu$ mol TE) per equivalents (TE) per kg of honey sample,  
189 respectively.

190  
191  
192

## 2.6. HPAEC/PAD analysis of sugars and sugar alcohols

193       The honey samples were homogenized, weighed (between 0.2 and 0.3 g) and diluted  
194       1000-fold with ultrapure water. The solutions were filtered and transferred to vials.

195       The sugar and sugar alcohol contents were determined by HPAEC/PAD.high  
196 ~~performance anion exchange chromatography with pulse amperometric detection~~  
197 (HPAEC/PAD). The honeys were analyzed on an ICS 3000 DP liquid chromatograph  
198 equipped with a quaternary gradient pump (Dionex, Sunnyvale, CA, USA). The  
199 carbohydrates were separated on a CarboPac®PA10 pellicular anion-exchange column (4 ×  
200 250 mm) at 30 °C. Each honey sample (25 µL) was injected with an ICS AS-DV 50  
201 autosampler (Dionex, Sunnyvale, CA, USA). The carbohydrates were eluted with the flow  
202 rate set to 0.7 mL/min, using a gradient program constituted from 600 mM sodium hydroxide  
203 (eluent A), 500 mM sodium acetate (eluent B) and ultrapure water (eluent C). The gradient  
204 program was as follows: 0.0–20.0 min, 15 % A; 20.1–30.0 min, 20 % A; 0.0–5.0 min, 0 % B;  
205 5.1–12.0 min, 2 % B; 12.1–20.0 min, 4 % B; 20.1–30.0 min, 20 % B, 0.0–5.0 min, 85 % C;  
206 5.1–12.0 min, 83 % C; 12.1–20.0 min, 81 % C; 20.1–30.0 min, 60 % C.Under these  
207 chromatographic conditions, the last compound was detected after approximately 25 min, and  
208 the analysis was ended at 30 min. The total amounts of each sugar or sugar alcohol was were  
209 evaluated according to the method previously described in section 2.3.

210

## 2.7. ICP-EOS analysis of minerals in honey samples

211       To analyze the mineral composition of honey, about 0.6–0.7 g of fresh honey sample  
212 was were treated with 7 mL of 65 % HNO<sub>3</sub> and 1 mL of 35 % H<sub>2</sub>O<sub>2</sub> in  
213 polytetrafluoroethylene (PTFE) vessels. A microwave closed digestion system (ETHOS 1,-;

215 Milestone, Bergamo, Italy) was used for the mineralization process. The final clear solution  
216 was made up to 50 mL with ultrapure water. A blank was prepared in the same way.

217 All mineral elements in the digested solutions were determined using an ICP-OES  
218 (iCAP 6500 Duo ICP, Thermo Scientific, UK) instrument. The results are expressed as mg of  
219 mineral metal per kg of honey.

220

221 *2.8. Statistical analysis*

222 Data of all measurements performed in triplicate are expressed as the mean ± standard  
223 deviation (SD). Statistical analyses were performed using the Analysis ToolPak from the  
224 Microsoft Office Excel 2007 Professional. Statistical analyses were performed with the  
225 program MS Excel (Microsoft Office 2007 Professional). PCA was realized using the PLS-  
226 Tool Box software package for MATLAB 7.12.0 (Eigenvector Research, Inc., Wenatchee,  
227 WA, USA) MATLAB (Version 7.12.0). All data were group-scaled prior to PCA. The  
228 singular value decomposition algorithm (SVD) and a 0.95 confidence level for  $Q$  and  $T^2$   
229 Hotelling limits for outliers were chosen.

230

231 **3. Results and discussion**

232

233 *3.1. Verification of the sage honey samples*

234 A great deal of attention was given to the authenticity of the Croatian Common sage  
235 honey samples, especially to their geographical and botanical origin (Persano Oddo and  
236 Bogdanov, 2004). Representative honey sampling was realized directly at primary producers'  
237 filling facilities, above all taking into the consideration two important criteria: A) that the  
238 honey sample extraction occurred closely soon after the sage flowering period (May) when  
239 sage flowers were the main bee source of nectar, and B) appropriate apiary locations for

240 sample production. That is to say, particular beehive sites were selected for collection of *S.*  
241 *officinalis* honey samples in line with the field observations on the abundance of sage nectar.  
242 Furthermore, cartographic data concerning the areas of predominate *Salvia officinalis* L.  
243 growth were taken from the comprehensive Vegetation-vegetation maps of Croatia (Šugar et  
244 al., 1983), confirming that the production beehives involved were situated deeply inside  
245 within the sage-dominated vegetation zones.

246 As a general rule, honey is considered unifloral if it was-is produced mainly from one  
247 plant species, and if the pollen of that particular species predominates. However, the pollen  
248 grains of some flowers are under-represented (or over-represented) in unifloral honeys, *i.e.*,  
249 the percentage of pollen in the sediment is lower (or higher) than the percentage of the  
250 corresponding nectar in the honey (Persano Oddo and Bogdanov, 2004). Therefore, the  
251 pollen spectrum of other nectariferous and non-nectariferous botanical species should  
252 likewise be taken into the consideration, as well as the presence of honeydew elements  
253 (Persano Oddo and Bogdanov, 2004; Piazza and Persano Oddo, 2004). The unifloral  
254 character of all the sage honey samples in this study was confirmed by thorough  
255 melissopalynological and sensory evaluation (**Table 1**). When compared to the representation  
256 of other pollen sources in samples, under-representation of *S. officinalis* pollen grains was  
257 noted in almost all the studied honey samples, thereby confirming the natural hypopollenic  
258 features of sage (Ricciardelli D'Albore and Galarini, 2000; Flora Croatica Database, 2012).  
259 The greatest-highest portion of the identified pollen in the sage unifloral honey originated  
260 from nectariferous species belonging to the families *Rhamnaceae*, *Sapindaceae* (genus *Acer*)  
261 and *Fagaceae* (genus *Castanea*). Pollen sources of non-nectariferous producing plants were  
262 mostly attributed to *Quercus* spp. (fam.-ily *Fagaceae*) and species belonging to the families  
263 *Graminaceae* and *Plantaginaceae* (*Plantago* spp.), all sharing the flowering period of sage as  
264 well as their areal of distribution. This characteristic pollen profile and specific combination

265 could be considered a valuable indicator of the geographical origin of the sage unifloral  
266 honey samples.

267 Sensory assessment, as an equally important analytical mechanism for the  
268 determination of the unifloral character of honey (Piana et al., 2004) revealed distinctive  
269 organoleptic features (visual, taste, odour, tactility) of the samples, taking into consideration  
270 the extent of their compliance with the particular organoleptic profile of unifloral sage honey  
271 (Lušić et al., 2007). Based on the results of the melissopalynological and sensory evaluations,  
272 all the honey samples in the present study were confirmed to be sage honeys.

273

### 274 *3.2. Phenolic profile of Croatian sage honey samples*

275 Although the composition of honey highly depends on the floral source used to collect  
276 the nectar, some other factors, including geographic origin, seasonal and environmental  
277 factors, bee variety, as well as processing technologies, may also affect the composition of  
278 the phenolic compounds in honey (Kaskonienė and Venskutonis, 2010). On the other hand,  
279 unifloral honeys have almost never been made from 100 % monofloral nectar, since the  
280 nectar from flowers of many various plants contributes to the production of every honey  
281 (Persano Oddo and Bogdanov, 2004). Therefore, it was important to analyze a large number  
282 of sage honey samples, in order to derive more general rules, and define which compounds  
283 and/or groups of compounds mostly characterize the phenolic and sugar profiles, and thus the  
284 uniqueness of this autochthonous honey. Phenolic compounds such as flavonoids (Kenjerić et  
285 al., 2008), carbohydrates (Primorac et al., 2011), and volatile compounds (Jerković et al.,  
286 2006), were previously suggested as possible markers for the determination of Common sage  
287 unifloral honey.

288 As it was previously reported, sage leaf extracts contain a wide range of phenolic compounds  
289 with the majority of the phenolic acids represented by caffeic acid derivatives and rosmarinic

acid being the dominant one. Sage leaf extracts, as hitherto reported, contain a wide range of phenolic compounds. The majority of the phenolic acids were found to be caffeic acid derivatives, with rosmarinic acid being the dominant one. According to Generalić et al. (2011), identified rosmarinic, syringic, gallic, *p*-coumaric, caffeic, and *trans*-ferulic acid as the principal phenolic acids of Common sage extracts. The principal phenolic acids of Common sage extracts are rosmarinic, syringic, gallic, *p*-coumaric, caffeic, and *trans*-ferulic acid. The relative content of rosmarinic acid in the extracts ranged from 94.54-% to 98.38-%, depending on the phenophase, while the contents of other acids were significantly lower (Generalić et al., 2011; Generalié et al., 2012). Other studies also report the presence of vanillic acid, salvianolic acids K and I, and methyl rosmarinate in Common sage (Dragovic-Uzelac et al., 2012; Dent et al., 2013). Flavonoids of *S. officinalis* are mostly present as flavones (apigenin, luteolin and their corresponding 6-hydroxylated derivatives), flavone glucosides (6-hydroxyluteolin-7-glucoside, luteolin-7-glucuronide, luteolin-glucoside, luteolin-3'-glucuronide, apigenin-7-glucuronide and apigenin-7-glucoside), flavonols (mostly kaempferol and quercetin methyl ethers), and flavonol glucosides (quercetin-4'-glucoside, rutin), as reported by several authors (Generalić et al., 2011; Dragovic-Uzelac et al., 2012; Generalić et al., 2012). Stilbenes (*trans*-resveratrol, astragin, piceid) and catechins ((+)-catechin, (-)-epicatechin) are also present (Generalić et al., 2011; Generalié et al., 2012). *Salvia officinalis* L. is reported to contain also phenolic diterpenes, including carnosol and carnosic acid (Lamien-Meda et al., 2010). Some of the phenolic compounds previously determined as constituents of sage leaf extracts were also found in unifloral *S. officinalis* honeys from this region (Kenjerić et al., 2008), including phenolic acids (caffeic, rosmarinic, gallic, *p*-coumaric, and ferulic acid), and flavonoids (flavones apigenin and luteolin, and their corresponding glycosides; flavonols quercetin and kaempferol and their derivatives (quercetin hexoside and rutin); stilbenes (resveratrol); and

315 catechins (catechin and epicatechin)). It should be borne in mind that previous studies  
316 concerning the composition of the phenolics in indigenous Croatian Common sage honey  
317 (Kenjerić et al., 2008) usually concentrated on targeted metabolomic analysis that included a  
318 limited number of compounds, and that there ~~is~~are a lack of literature data concerning the  
319 complete polyphenolic profiles.

320 On the other hand, the present study gives insight into the profile of the phenolics of sage  
321 unifloral honey using the non-targeted metabolomic approach, which resulted in the  
322 identification of a significant number of phenolic compounds (**Table 2**). In the absence of  
323 standards, the identification of flavonoid glycosides and other phenolics were based on the  
324 search for the  $[M-H]^-$  deprotonated molecule and its fragmentation using UHPLC-LTQ  
325 OrbiTrap MS/MS. The exact mass search and the study of the fragmentation pathways  
326 described in the literature enabled as much structural information as possible to be obtained.

327 In this way, it was possible to ~~individuate~~identify 61 compounds (**Table 2**). The  
328 chromatograms of the investigated Common sage honey samples showed similar profiles. A  
329 selected base peak chromatogram of a representative sage honey extract (sample No. SH2) is  
330 shown in **Fig. S1**.

331 Hydroxycinnamic acids, such as caffeic, rosmarinic, ferulic, chlorogenic, and *p*-  
332 coumaric acid were detected in the sage honey samples analyzed in the present study. These  
333 phenolic acids constituted a significant share ~~to~~of the total phenolics content of the sage  
334 honey samples. Generally, the presence of phenolic compounds in nectar is usually connected  
335 with their protective role against microbial infestations (Heil, 2011). However, high  
336 concentrations of these compounds could lead to ~~the~~the nectar's toxic effect, and have a negative  
337 influence on pollinators (Adler, 2000). Of the hydroxybenzoic acids, *p*-hydroxybenzoic acid,  
338 vanillic, gentisic, and protocatechuic acid were previously reported in sage (Zgórska and  
339 Główniak, 2001), and confirmed in unifloral sage honey samples. All these phenolic acids are

340 considered as potential markers for the authentication of sage unifloral honeys and were  
341 therefore included in the subsequent targeted quantitative analyses of the honey samples.

342 The majority of flavonoids in *S. officinalis* are flavones of apigenin and luteolin, and  
343 their corresponding 6-hydroxylated derivatives (hispidulin and cirsimarin), as well as the  
344 dihydroflavone hesperetin (Brieskorn and Biechele, 1971; Cuvelier et al., 1996; Lu and Yeap  
345 Foo, 2002; Kontogianni et al., 2013), and all of these compounds were evidenced in the  
346 analyzed sage honey samples. Of the flavone, glucosides, luteolin and apigenin glycosides  
347 are very common in analyzed sage honeys, and some of them were previously found in *S.*  
348 *officinalis* (Masterova et al., 1989; Wang et al., 1998; Lu and Yeap Foo, 2000). Interestingly,  
349 it is well known that the presence of 6-hydroxy- and 6-methoxy-flavone glycosides clearly  
350 differentiates section *Salvia*, which includes *S. officinalis*, from other sections belonging to  
351 the genus *Salvia* (Tomás-Barberán et al., 1988). Therefore, the presence of these compounds  
352 in honey might be one of the indicators that the honey in question is really of sage floral  
353 origin. Flavonols of sage are mostly those of kaempferol and quercetin methyl ethers (Lu and  
354 Yeap Foo, 2002), and nectar-pollen derived flavonoids, such as quercetin, kaempferol,  
355 and hesperetin, have been identified in samples of Common sage honey. Of the flavonoids  
356 previously identified in sage, stilbene resveratrol and catechins (catechin and epicatechin)  
357 were also confirmed in the sage honey samples (Generalić et al., 2011). The following  
358 derivatives of catechin and epicatechin were also recorded in the honey samples:  
359 gallicatechin, epigallicatechin, gallicatechin gallate, and epigallicatechin gallate.

360 The phenolic diterpenes carnosol and carnosic acid, although present in sage  
361 (Kontogianni et al., 2013), were not previously detected in unifloral sage honeys. In the  
362 present study, these compounds were identified in the all honey samples, but in trace amounts  
363 (**Table 2**).

364

## 365 3.3. Quantification of targeted phenolics in the honey samples

366 Solid-Solid-phase extraction (SPE) combined with ultra-high-performance liquid  
367 chromatography with a diode array detector (DAD) and a triple-quadrupole mass  
368 spectrometer was used to analyze the content of 25 targeted compounds in the *S. officinalis*  
369 honey samples. Three basic criteria for the selection of chemical markers from the group of  
370 phenolic compounds were applied: 1) putative sage nectar-pollen derived compounds  
371 (phenolic acids and flavonoids); 2) propolis characteristic flavonoids and 3) abscisic acid.

372 Among the quantified compounds in Common sage honeys, some of phenolic acids,  
373 *i.e.*, *p*-coumaric, *p*-hydroxybenzoic, and ferulic acid, were present in the highest amounts.

374 Interestingly, rosmarinic acid was present in relatively low amounts in the unifloral sage  
375 honeys analyzed in the present study (**Table 3**). It is well known that phenolic acids of sage

376 are mostly based on caffeic acid building blocks (Lu and Yeap Foo, 2002), and that

377 rosmarinic acid is the major phenolic compound in sage leaves. Possible reasons for this  
378 could be relatively low concentrations of this compound in the nectar. Gentisic acid was

379 detected only in three samples (SH8, SH15, and SH18). Of the nectar-pollen derived  
380 flavonoids quantified herein, quercetin, kaempferol, and hesperetin were abundant and

381 present in significant amounts. Stilbene resveratrol was detected only in four of the sage

382 honey samples (SH2, SH5, SH16, and SH17). Catechins were abundant in the analyzed  
383 honey samples, with gallocatechin gallate and epigallocatechin gallate being quantified as the  
384 dominant compounds from this group. The contents of catechin and epicatechin were low.

385 Pinocembrin, pinobanksin, pinostrobin, galangin, and chrysin are characteristic  
386 flavonoids of propolis, and were determined in most of the previously analyzed European  
387 honey samples (Tomás-Barberán et al., 2001; Kenjerić et al., 2008). The portion of propolis-  
388 derived compounds in the unifloral sage honeys analyzed in the present study was significant,  
389 but much less than in a previous study (Kenjerić et al., 2008), which reported a relatively

390 high portion of galangin and chrysin (51.3-%) in the total identified flavonoids. The sage  
391 honey samples analyzed in the present study were characterized by the significant amounts of  
392 pinobaksin (0.21–2.35 mg/kg) and chrysin (0.06–1.98 mg/kg).

393 The plant stress hormone abscisic acid (AbBA) is known to be present in floral  
394 nectars of some plants, and is transferred from the nectar to honey. This phytohormone is  
395 present in relatively high amounts in some European honeys (Tomás-Barberán et al., 2001;  
396 Truchado et al., 2008; Bertoncelj et al., 2011), including unifloral sage honey (Kenjerić et al.,  
397 2008), and was also confirmed in the present study. The presence of abscisic acid in high  
398 amounts (0.26–3.99 mg/kg) is not surprising, since natural rocky habitat of sage is  
399 characterized by periods of drought seasons during the summer, which results in stress-  
400 induced responses in the plants (Bertoncelj et al., 2011).

401

#### 402 3.4. Antioxidant activity of Common sage honeys

403 Antioxidant capacity of *S. officinalis* honey samples was determined by the total  
404 phenolics content (TPC) and the radical scavenging activity (RSA). The results of these  
405 investigations are given in **Table 3**.

406 The Common sage honey samples were characterized with TPC values ranging  
407 between 208.519 to and 747.549 mg of gallie acid equivalents (GAE) per kg of honey. The  
408 average content of total phenolics was in a good agreement with the values given in the  
409 literature for sage honeys from the same region (Piljac-Žegarac et al., 2009).

410 The results of the determination of the RSA of sage honey samples ranged from  
411 351.20 to 894.8275 micromoles of Trolox equivalents TE per kg of sample. To determine the  
412 relationship between the content of polyphenols and antioxidant activities of *S. officinalis*  
413 honey samples, the correlation between the TPC and the RSA values was calculated. The  
414 RSA showed a statistically significant ( $r = 0.872$ ;  $P_p < 0.0001$ ) and positive linear

415 correlation with the TPC ( $RSA = 68.08 + 1.10 \times TPC$ ). A significant and positive linear  
416 relationship between the antioxidant activity and total phenolic content of sage honey  
417 samples indicated that phenolic compounds could be identified as the chemicals that  
418 predominately contributed to the antioxidant activity, which is in accordance with previous  
419 investigations ~~reported previously~~ (Piljac-Žegarac et al., 2009; Gasic et al., 2014).

420

421 *3.5. Determination of the sugars and sugar alcohols*

422 Fourteen different sugars and sugar alcohols were identified and quantified in the  
423 analyzed unifloral sage honey samples using the HPAEC/PAD-method. Quantification was  
424 performed with available standards. The reducing sugars, fructose and glucose, were found to  
425 be the major constituents of all the investigated samples (**Table 4**), which confirmed that all  
426 honey samples were genuine honeys. In all the analyzed honeys, the value of the glucose plus  
427 fructose amounts was around or higher than 60g per 100 g, which is the value for all honey  
428 types required by the European and FAO (Codex Alimentarius) standards (FAO/WHO, 2001;  
429 The Council of the European Union, 2002). Another monosaccharide identified in the honeys  
430 in relatively low amounts was arabinose.

431 All the sage honey samples had a sucrose content lower than 5g per 100 g, which is  
432 generally taken as the limit value for honeys allowed by European Union Honey Directive  
433 (The Council of the European Union, 2002). Apart from sucrose, the other identified  
434 disaccharides were trehalose, turanose, maltose and isomaltose. The trisaccharides  
435 maltotriose and isomaltotriose were also evidenced. From the group of polyols (sugar  
436 alcohols), erythritol, sorbitol, ~~galactitol~~galactitol, and glycerol were identified.

437 The ratio between some carbohydrates is another indicator that may be used to  
438 ascertain honey authenticity. Thus, the ratios of fructose/glucose, maltose/isomaltose,  
439 sucrose/turanose, and maltose/turanose, maltotriose/raffinose+erlose+melezitose were used

440 for the authentication of some unifloral honeys, and all these studies were reviewed by  
441 Kaskonienė and Venskutonis (2010). The fructose/glucose (*FRU/GLU*) ratio in sage honeys,  
442 which was recommended for the evaluation of honey granulation because glucose is less  
443 ~~water~~-soluble than fructose, varied from 1.31 in sample **SH1** to 4.42 in sample **SH87**.  
444 One more characteristic of the unifloral sage honeys analyzed in the present study was the  
445 relatively low maltose/isomaltose (*MAL/iMAL*) ratio, which ranged from 0.9 (sample **SH11**)  
446 to 2.41 (**SH5**).  
447

#### 448 3.6. Determination of minerals in *S. officinalis* honeys

449 The concentrations of minerals quantified in the studied sage honey samples are  
450 presented in **Table 5**. The most abundant element in all samples was found to be potassium  
451 (content ranging from 5921.68 to 2151.350 mg/kg), which agrees with other studies and  
452 indicates that K is the most common element in honeys (Cantarelli et al., 2008), including  
453 unifloral sage honeys (Bilandžić et al., 2014). Phosphorus, sulfur, and calcium were the next  
454 most common elements, followed by magnesium and sodium. Among the micro-elements in  
455 decreasing amounts, B, Zn, Fe, Mn, Cu, Se, and Ni were found, while Co, Cr, Li, and V were  
456 found as trace elements. Therefore, the influence of botanical origin on the elemental  
457 composition of the unifloral sage honey was evident for both elements essential for plant  
458 growth (macronutrients), such as K, P, S, Ca, Mg and Na, and for micronutrients (trace  
459 elements), such as B, Mn, Zn, Fe, etc. The essential elements are present in plants in  
460 significantly higher amounts than the trace elements, and this observation was also true for  
461 the honey samples. On the other hand, the possibility that the mineral composition of honey  
462 samples also reflects the environmental and pedological conditions of the geographical  
463 locality cannot be excluded (Terrab et al., 2004). Toxic elements (Al, As, Cd, Pb, and Sb) in

464 the tested samples were found in small amounts (allowable concentrations), which excludes  
465 the existence of environmental contamination of the honeys.

466

467 *3.7. Pearson's correlation analysis*

468 Pearson's correlation analysis was performed to evaluate the associations between  
469 variables in 18 sage unifloral honey samples (**Table S2**), in order to define some general rules  
470 characteristic for unifloral sage honey. Both positive and negative Pearson's correlations  
471 were observed between the contents of the different analyzed compounds in the unifloral sage  
472 honeys. However, statistically significant correlations were observed in some cases as can be  
473 seen from the Tables given in supplementary material (**Tables S2–S4**). High positive  
474 correlations were found between propolis-derived compounds. Namely, correlations between  
475 CaA and PNB, PNS, CHR, PNC, GLN were in the range from 0.691 to 0.886. Likewise,  
476 correlations among PNB, PNS, CHR, PNC, and GLN were also characterized with high  
477 positive coefficients (**Table S2**). Statistically significant correlations between CaA and HES  
478 ( $r = 0.827, P_{p \leq} 0.0005$ ), FeA and GeA ( $r = 0.786, P_{p \leq} 0.0005$ ), FeA and PrA ( $r = 0.652, P_{p \leq} 0.005$ ),  
479 C and EC ( $r = 0.663, P_{p \leq} 0.005$ ), and C and EGC ( $r = 0.656, P_{p \leq} 0.005$ ) could  
480 be considered as important characteristics of the analyzed sage honeys. It was also observed  
481 that AbA was well correlated with FeA and GeA, with  $r = 0.890 (P_{p \leq} 0.000001)$  and  $r = 0.887 (P_{p \leq} 0.000001)$ , respectively. The observed correlations between the phenolic  
482 compounds in the analyzed honey samples probably reflected the situation in the sage nectar  
483 and/or pollen, which are the main sources of phenolics in honey.

485 Pearson's correlation analysis was also performed between 14 targeted carbohydrates  
486 in the unifloral sage honey samples (**Table S3**), whereby the highest positive correlation was  
487 observed between maltose and isomaltose ( $r = 0.870, P_{p \leq} 0.000005$ ), which could be  
488 considered as a unique characteristic of unifloral sage honey. Moreover, statistically

489 significant correlations were found between MALT and SUC, and MALT and TURmaltotriose  
 490 and sucrose, and maltotriose and turanose (**Table S3**).

491 Regarding the mineral composition of the sage honeys, among all statistically  
 492 significant correlations, the highest positive ones were between Ca and Mn with  $r = 0.858$  ( $P$   
 493  $p \leq 0.000005$ ), and between Mg and P with  $r = 0.849$  ( $P p \leq 0.000005$ ). **Table S4** shows the  
 494 Pearson's correlation analysis of the minerals.

495

#### 496 3.8. Authentication of unifloral sage honey

497 In order to demonstrate the applicability of the present research for the authentication  
 498 of unifloral sage honey, three types of available unifloral honeys of *Lamiaceae* species were  
 499 introduced into the analysis as out-groups: mint (*Mentha* spp.) honey, winter savory (*Satureja*  
 500 *montana* L.) honey, and thyme (*Thymus* spp.) honey. The quantitative data on TPC, RSA,  
 501 targeted phenolics, sugars and minerals in thyme, mint and winter savory honeys are  
 502 presented as Supplementary data (**Table S5**). Principal component analysis (PCA) was  
 503 employed to analyze the quantitative data for TPC, RSA, 25 targeted phenolic compounds, 14  
 504 carbohydrates and 25 minerals in order to examine their relative variations within different  
 505 honeys (sage, mint, thyme and winter savory honeys).

506 The combination of all the variables was informative enough to clearly discriminate  
 507 sage honeys from the honeys of different floral origins. The results showed that the principal  
 508 factorial 2-dimensional plane captured 32.18-% of the total variability (**Fig. 1**). The first  
 509 principal component accounted for 17.58-% and the second for 15.60-% of the total variance.  
 510 Clear differentiation of unifloral sage honey from unifloral thyme, mint and winter savory  
 511 honeys along PC 1 was observed. The variables responsible for the differentiation of unifloral  
 512 sage honey from the other studied honeys were identified using the loading plots (**Fig. 1B**).  
 513 Sage honey samples were distinguished from the other studied honeys based on the

514 significantly higher contents of mineral-boron-B. Most of the samples of sage honeys were  
515 characterized with high K contents. Higher contents of TPC, ~~TUR~~-turanose and ~~KAE~~  
516 kaempferol in the sage honeys compared to the thyme, mint and winter savory honeys further  
517 contributed to the separation (Fig. 1B). On the other hand, mint honey was characterized by  
518 larger contents of Mn, Ba, and ~~ChA~~chlorogenic acid, when compared to the other samples.  
519 Only two unifloral sage samples (**SH2** and **SH8**) considerably deviated from the rest of the  
520 sage honey samples along PC2, due to higher contents of chrysanthemic acid, pinocembrin,  
521 galangin~~CHR~~, PNC, GLN, and ~~CaA~~caffeic acid, which were also characteristic for the **MH1**  
522 and **WSH2** samples.

523

#### 524 4. Conclusions

525

526 The study of sage (*Salvia officinalis* L.) honey samples showed somewhat interesting  
527 results related to their peculiar-characteristic phenolic, sugar and mineral contents. Several  
528 identified compounds showed significant potential for the characterization of this particular  
529 honey intrinsic-fortypical of the Adriatic Littoral of Croatia, especially its northern area. The  
530 data suggest clear differentiation of unifloral sage honey from the other unifloral honeys by  
531 using groups of chemical markers (phenolic compounds, carbohydrates and minerals).

532 Among all studied unifloral honeys of *Lamiaceae* species, higher contents of boron and  
533 potassium, as well as turanose and kaempferol could be identified as authentication markers  
534 of unifloral sage honey. In addition, the application of multivariate statistical analysis ~~to~~-for  
535 the authentication and classification ~~was~~ proved to be an important complementary tool for a  
536 more reliable identification and quality control method of honey.

537

538

539

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545

546 **References**

547

548 Adler, L.S., (2000). The ecological significance of toxic nectar. *Oikos* 91(3), 409-420.

549 Bertoncelj, J., Polak, T., Kropf, U., Korošec, M., Golob, T., (2011). LC-DAD-ESI/MS  
550 analysis of flavonoids and abscisic acid with chemometric approach for the classification of  
551 Slovenian honey. *Food Chemistry* 127(1), 296-302.

552 Bilandžić, N., Gačić, M., Đokić, M., Sedak, M., Šipušić, Đ.I., Končurat, A., Gajger, I.T.,  
553 (2014). Major and trace elements levels in multifloral and unifloral honeys in Croatia. *Journal*  
554 of Food Composition and Analysis

555 33(2), 132-138.  
556 Brieskorn, C.H., Biechele, W., (1971). Flavones from *Salvia officinalis*. Compounds of  
557 *Salvia officinalis*. *Archiv der Pharmazie* (Weinheim) 304, 557–561.

558 Cantarelli, M.A., Pellerano, R.G., Marchevsky, E.J., Camiña, J.M., (2008). Quality of honey  
559 from Argentina: study of chemical composition and trace elements. *The Journal of the*  
Argentine Chemical Society 96, 33-41.

560 Cuvelier, M.-E., Richard, H., Berset, C., (1996). Antioxidative activity and phenolic  
561 composition of pilot-plant and commercial extracts of sage and rosemary. *Journal of the*  
562 American Oil Chemists' Society 73(5), 645-652.

563 Dent, M., Dragovic-Uzelac, V., Penić, M., Brncic, M., Tomislav, B., Levaj, B., (2013). The  
564 Effect of Extraction Solvents, Temperature and Time on the Composition and Mass Fraction  
565 of Polyphenols in Dalmatian Wild Sage (*Salvia officinalis* L.) Extracts. *Food Technology &*  
566 *Biotechnology* 51(1), 84–91.

567 Dragovic-Uzelac, V., Elez Garofulić, I., Jukić, M., Penić, M., Dent, M., (2012). The  
568 Influence of Microwave-Assisted Extraction on the Isolation of Sage (*Salvia officinalis* L.)  
569 Polyphenols. *Food Technology & Biotechnology* 50(3), 377–383.

- 570 FAO/WHO, (2001). Codex standards for honey, in: Codex Alimentarius Commission (Ed.),  
571 2nd revision. Food and Agriculture Organisation of the United Nations, World Health  
572 Organisation, Rome, pp. 1-8.
- 573 Flora Croatica Database, (2012). Flora Croatica Database. Vascular Plants, Taxonomy &  
574 Bibliography of Croatian Flora, 2004 ed. Department of Botany, Faculty of science , FER-  
575 ZPR , University of Zagreb.
- 576 Gasic, U., Keckes, S., Dabic, D., Trifkovic, J., Milojkovic-Opsenica, D., Natic, M., Tesic, Z.,  
577 (2014). Phenolic profile and antioxidant activity of Serbian polyfloral honeys. Food  
578 Chemistry 145, 599-607.
- 579 Generalić, I., Skroza, D., Ljubenkov, I., Katalinić, A., Burčul, F., Katalinić, V., (2011).  
580 Influence of the phenophase on the phenolic profile and antioxidant properties of Dalmatian  
581 sage. Food Chemistry 127(2), 427-433.
- 582 Generalić, I., Skroza, D., Šurjak, J., Mozina, S.S., Ljubenkov, I., Katalinić, A., Simat, V.,  
583 Katalinić, V., (2012). Seasonal variations of phenolic compounds and biological properties in  
584 sage (*Salvia officinalis* L.). Chemistry & Biodiversity 9(2), 441-457.
- 585 Heil, M., (2011). Nectar: generation, regulation and ecological functions. Trends in Plant  
586 Science 16(4), 191-200.
- 587 Jerković, I., Mastelić, J., Marijanović, Z., (2006). A Variety of Volatile Compounds as  
588 Markers in Unifloral Honey from Dalmatian Sage (*Salvia officinalis* L.). Chemistry &  
589 Biodiversity 3, 1307-1310.
- 590 Karousou, R., Hanlidou, E., Kokkini, S., (2000). The sage plant of Greece: distribution and  
591 intraspecific variation, in: Kintzios, S.E. (Ed.), *Sage the genus Salvia*. Harwood Academic  
592 Publishers, Amsterdam, pp. 27–46.

- 593 Kaskonienė, V., Venskutonis, P.R., (2010). Floral Markers in Honey of Various Botanical  
594 and Geographic Origins: A Review. Comprehensive Reviews in Food Science and Food  
595 Safety 9(6), 620-634.
- 596 Kenjerić, D., Mandić, M., Primorac, L., Čačić, F., (2008). Flavonoid pattern of sage (*Salvia*  
597 *officinalis* L.) unifloral honey. Food Chemistry 110, 187-192.
- 598 Kontogianni, V.G., Tomic, G., Nikolic, I., Nerantzaki, A.A., Sayyad, N., Stosic-Grujicic, S.,  
599 Stojanovic, I., Gerothanassis, I.P., Tzakos, A.G., (2013). Phytochemical profile of  
600 Rosmarinus officinalis and Salvia officinalis extracts and correlation to their antioxidant and  
601 anti-proliferative activity. Food Chemistry 136(1), 120-129.
- 602 Lamien-Meda, A., Nell, M., Lohwasser, U., Borner, A., Franz, C., Novak, J., (2010).  
603 Investigation of antioxidant and rosmarinic acid variation in the sage collection of the  
604 genebank in Gatersleben. Journal of Agricultural and Food Chemistry 58(6), 3813-3819.
- 605 Loveaux, J., Maurizio, A., Vorwohl, G., (1978). Methods of Melissopalynology by  
606 International Commission for bee Botany or IUBS. Bee World 59(4), 139-157.
- 607 Lu, Y., Yeap Foo, L., (2000). Flavonoid and phenolic glycosides from *Salvia officinalis*.  
608 Phytochemistry 55(3), 263-267.
- 609 Lu, Y., Yeap Foo, L., (2002). Polyphenolics of *Salvia*—a review. Phytochemistry 59(2), 117-  
610 140.
- 611 Lušić, D., Koprivnjak, O., Ćurić, D., Sabatini, A., Conte, L., (2007). Volatile Profile of  
612 Croatian Lime Tree (*Tilia* sp.), Fir Honeydew (*Abies alba*) and Sage (*Salvia officinalis*)  
613 Honey. Food Technology & Biotechnology 45(2), 156-165.
- 614 Masterova, I., Uhrin, D., Kettmann, V., Suchy, V., (1989). Phyto chemical study of *Salvia*  
615 *officinalis*. Chemical Papers 43(6), 797—803.

- 616 Natic, M.M., Dabic, D.C., Papetti, A., Fotiric Aksic, M.M., Ognjanov, V., Ljubojevic, M.,  
617 Tesic, Z., (2015). Analysis and characterisation of phytochemicals in mulberry (*Morus alba*  
618 L.) fruits grown in Vojvodina, North Serbia. *Food Chem* 171, 128-136.
- 619 Patiny, L., Borel, A., (2013). ChemCalc: A Building Block for Tomorrow's Chemical  
620 Infrastructure. *Journal of Chemical Information and Modeling* 53(5), 1223-1228.
- 621 Persano Oddo, L., Bogdanov, S., (2004). Determination of honey botanical origin: problems  
622 and issues. *Apidologie* 35, S2-S3.
- 623 Piana, M., Persano Oddo, L., Bentabol, A., Bruneau, E., Bogdanov, S., Guyot Declerck, C.,  
624 (2004). Sensory analysis applied to honey: state of the art. *Apidologie* 35(Suppl. 1), S26-S37.
- 625 Piazza, M., Persano Oddo, L., (2004). Bibliographic review of main unifloral European  
626 honeys. *Apidologie* 35, S94-S111.
- 627 Piljac-Žegarac, J., Stipčević, T., & Belščak, A., (2009). Antioxidant properties and phenolic  
628 content of different floral origin honeys. *Journal of ApiProduct and ApiMedical Science* 1(2),  
629 43-50.
- 630 Primorac, L., Flanjak, I., Kenjerić, D., Bubalo, D., Topolnjak, Z., (2011). Specific Rotation  
631 and Carbohydrate Profile of Croatian Unifloral Honeys. *Czech Journal of Food Sciences*  
632 29(5), 515-519.
- 633 Ricciardelli D'Albore, G., Galarini, R., (2000). Mediterranean Melissopalynology. *Istituto*  
634 *Zooprofilattico Sperimentale dell'Umbria e delle Marche, Perugia Italy.*
- 635 Šugar, I., Gaži-Baskova, V., Trinajstić, I., Horvatić-Hodak, N., Lovrić, A., Horvatić, S.,  
636 Kutleša, L., (1983). *Vegetacijska karta SR Hrvatske*, in: Botanički zavod Prirodoslovno-  
637 matematički fakultet (Ed.). *Vojnogeografski institut Beograd, Zagreb.*
- 638 Terrab, A., Hernanz, D., Heredia, F.J., (2004). Inductively coupled plasma optical emission  
639 spectrometric determination of minerals in thyme honeys and their contribution to  
640 geographical discrimination. *J Agric Food Chem* 52(11), 3441-3445.

- 641 The Council of the European Union, (2002). Council Directive 2001/110/EC of 20 December  
642 2001 relating to honey, in: Union, T.C.o.t.E. (Ed.). Official Journal of the European  
643 Communities, pp. 47-49.
- 644 Tomás-Barberán, F.A., Grayer-Barkmeijer, R.J., Gil, M.I., Harborne, J.B., (1988).  
645 Distribution of 6-hydroxy-, 6-methoxy- and 8-hydroxyflavone glycosides in the labiateae, the  
646 scrophulariaceae and related families. Phytochemistry 27(8), 2631-2645.
- 647 Tomás-Barberán, F.A., Martos, I., Ferreres, F., Radovic, B.S., Anklam, E., (2001). HPLC  
648 flavonoid profiles as markers for the botanical origin of European unifloral honeys. Journal of  
649 the Science of Food and Agriculture 81(5), 485-496.
- 650 Truchado, P., Ferreres, F., Bortolotti, L., Sabatini, A.G., Tomas-Barberan, F.A., (2008).  
651 Nectar Flavonol rhamnosides are floral markers of acacia (*Robinia pseudacacia*) honey.  
652 Journal of Agricultural and Food Chemistry 56(19), 8815-8824.
- 653 Von der Ohe, W., Persano Oddo, L., Piana, M., Morlot, M., Martin, P., (2004). Harmonized  
654 methods of melissopalynology. Apidologie 35, S18-S25.
- 655 Wang, M., Li, J., Rangarajan, M., Shao, Y., LaVoie, E.J., Huang, T.-C., Ho, C.-T., (1998).  
656 Antioxidative Phenolic Compounds from Sage (*Salvia officinalis*). Journal of Agricultural  
657 and Food Chemistry 46(12), 4869-4873.
- 658 Zgórska, G., Głowniak, K., (2001). Variation of free phenolic acids in medicinal plants  
659 belonging to the Lamiaceae family. Journal of Pharmaceutical and Biomedical Analysis  
660 26(1), 79-87.
- 661

662           **Figure Captions**

663

664   **Fig. 1.** (A) PC scores plot of the honey samples; (B) Loadings plot of the honey samples.

665   **Fig. S1.** Base peak chromatogram of Common sage honey (sample No. SH2) extract. Peak

666           numbers corresponds to those in **Table 3**: (2) gallicatechin, (3) salvianic acid  $\alpha\Delta$ , (4)  
667           protocatechuic acid, (7) epigallocatechin, (9) catechin, (11) chlorogenic acid, (12) *p*-  
668           hydroxybenzoic acid, (13) feruloyl-hexoside, (14) epicatechin, (17) coumaroyl-  
669           hexoside, (22) gentistic acid, (23) luteolin-rutinoside, (24) isorhamnetin-rutinoside,  
670           (25) quercetin-hexoside, (27) *p*-coumaric acid, (28) taxifolin, (30) rosmarinic acid,  
671           (35) *trans, trans*-abscisic acid, (37) monohydroxybenzoic acid, (42) sakuranetin, (44)  
672           kaempferol, and (45) rhamnetin.

673

674

675 **Table 1** Apiary locations of the sage honey sample's production. Melissopalynological and  
 676 sensory assessment of unifloral sage (*Salvia officinalis* L.) honey samples deriving from the  
 677 North Croatian Littoral.

Sample	Location	Year	Melissopalynological	Sensory	Compliance to
			assessment of honey samples	assessment of honey samples	sage honey uniflormality
SH1	Croatia, Cres	2013	D	Fair	Complies
SH2	Croatia, Eastern Istria	2013	B	Good	Complies
SH3	Croatia, Cres	2013	C	Fair	Complies
SH4	Croatia, Rab	2012	C	Fair	Complies
SH5	Croatia, Cres	2012	C	Good	Complies
SH6	Croatia, Krk	2011	B	Good	Complies
SH7	Croatia, Klenovica	2011	C	Good	Complies
SH8	Croatia, Krk	2011	B	Fair	Complies
SH9	Croatia, Cres	2010	C	Good	Complies
SH10	Croatia, Krk	2010	B	Good	Complies
SH11	Croatia, Cres	2010	D	Good	Complies
SH12	Croatia, Krk	2010	C	Fair	Complies
SH13	Croatia, Krk	2009	C	Good	Complies
SH14	Croatia, Cres	2009	C	Fair	Complies
SH15	Croatia, Kraljevica	2012	B	Good	Complies
SH16	Croatia, Cres	2010	C	Fair	Complies
SH17	Croatia, Cres	2012	C	Fair	Complies
SH18	Croatia, Krk	2012	B	Good	Complies

678 Pollen frequency classes:

679 A - "Predominant pollen" (more than 45 % of the pollen grains);

680 B - "Secondary pollen" (16–45 %);

681 C - "Important minor pollen" (3–15 %);

682 D - "Minor pollen" (less than 3 %).

683

684 **Table 2** Presence of polyphenolics in the sage (*Salvia officinalis* L.) honeys; number of  
 685 identified compound, target compounds, mean expected retention times, exact mass,  
 686 calculated mass, mean mass accuracy (ppm), and MS/MS fragments.

Peak No	Compounds	<i>t</i> <sub>R</sub> , min	Exact mass, [M-H] <sup>-</sup>	Calculated mass [M-H] <sup>-</sup>	Δ pm	MS/MS fragments
1	Gallic acid <sup>a</sup>	2.55	169.01392	169.01425	1.95	125
2	Gallocatechin <sup>a</sup>	3.96	305.06583	305.06668	2.79	219, 261
3	Salvianic acid A	3.97	197.04520	197.04555	1.78	179, 153, 123
4	Protocatechuic acid <sup>a</sup>	4.34	153.01903	153.01933	1.96	109
5	Chlorogenic acid isomer 1	4.48	353.08716	353.08781	1.84	191, 179, 146
6	Caffeoyl-hexoside	4.52	341.08716	341.08781	1.91	179
7	Epigallocatechin <sup>a</sup>	4.65	305.06589	305.06668	2.59	219, 261
8	Dimethoxybenzoic acid	4.75	181.05025	181.05063	2.10	151, 137
9	Catechin <sup>a</sup>	4.92	289.07095	289.07176	2.80	159, 123
10	Eriodictyol-rutinoside	5.01	595.16644	595.16684	0.67	449, 287
11	Chlorogenic acid <sup>a</sup>	5.04	353.08682	353.08781	2.80	191, 179, 146
12	p-Hydroxybenzoic acid <sup>a</sup>	5.09	137.02425	137.02442	1.24	93
13	Feruloyl-hexoside isomer 1	5.30	355.10229	355.10346	3.29	193
14	Epicatechin <sup>a</sup>	5.32	289.07114	289.07176	2.14	159, 123
15	Gallocatechin gallate <sup>a</sup>	5.34	457.07703	457.07763	1.31	305
16	Chlorogenic acid isomer 2	5.37	353.08710	353.08781	2.01	191, 179, 146
17	Coumaroyl-hexoside	5.39	325.09213	325.09289	2.34	163
18	Epigallocatechin gallate <sup>a</sup>	5.46	457.0769	457.07763	1.60	305
19	Caffeic acid <sup>a</sup>	5.48	179.03476	179.03498	1.23	135, 161
20	Feruloyl-hexoside isomer 2	5.61	355.10260	355.10346	2.42	193
21	Rutin <sup>a</sup>	5.94	609.14490	609.14611	1.99	463, 301
22	Gentistic acid <sup>a</sup>	5.96	153.01900	153.01933	2.16	109
23	Luteolin-rutinoside	5.97	593.15045	593.15119	1.25	447, 285
24	Isorhamnetin-rutinoside	6.03	623.16040	623.16176	2.18	461, 315
25	Quercetin-hexoside	6.07	463.08691	463.08820	2.79	301
26	Ellagic acid <sup>a</sup>	6.16	300.99847	300.99899	1.73	283, 200, 175
27	p-Coumaric acid <sup>a</sup>	6.25	163.03984	163.04007	1.41	119

<b>28</b>	<b>Taxifolin</b>	6.47	303.05023	303.05103	2.64	285, 269, 255, 217
<b>29</b>	<b>Ferulic acid<sup>a</sup></b>	6.70	193.05014	193.05063	2.54	175, 139
<b>30</b>	<b>Rosmarinic acid<sup>a</sup></b>	6.79	359.07635	359.07724	2.48	197, 179, 161
<b>31</b>	<b>Apigenin-rutinoside isomer</b>	6.83	577.15521	577.15628	1.85	431, 269
<b>32</b>	<b>Apigenin-hexoside isomer 1</b>	6.92	431.09775	431.09837	1.44	269
<b>33</b>	<b>Luteolin-hexoside</b>	6.93	447.09274	447.09329	1.23	285
<b>34</b>	<b>Eriodictyol</b>	7.14	287.05551	287.05611	2.09	125
<b>35</b>	<b><i>trans, trans</i>-Abscisic acid</b>	7.44	263.12814	263.12888	2.81	191, 179
<b>36</b>	<b>Apigenin-hexoside isomer 2</b>	7.52	431.09778	431.09837	1.37	269
<b>37</b>	<b>Monohydroxybenzoic acid</b>	7.70	137.02423	137.02442	1.39	93
<b>38</b>	<b><i>cis, trans</i>-Abscisic acid<sup>a</sup></b>	7.73	263.12833	263.12888	2.09	191, 179
<b>39</b>	<b>Luteolin<sup>a</sup></b>	7.75	285.03989	285.04046	2.00	213, 151
<b>40</b>	<b>Quercetin<sup>a</sup></b>	7.80	301.03445	301.03538	3.09	151, 179, 121
<b>41</b>	<b>Resveratrol<sup>a</sup></b>	7.85	227.07056	227.07137	3.57	209
<b>42</b>	<b>Sakuranetin</b>	8.06	285.07623	285.07685	2.17	133
<b>43</b>	<b>Apigenin<sup>a</sup></b>	8.44	269.04477	269.04555	2.90	149, 151, 173, 183
<b>44</b>	<b>Kaempferol<sup>a</sup></b>	8.57	285.03970	285.04046	2.67	199, 161, 151, 135
<b>45</b>	<b>Rhamnetin</b>	8.57	315.04996	315.05103	3.40	300, 165, 121
<b>46</b>	<b>Hispidulin</b>	8.66	299.05527	299.05611	2.81	284
<b>47</b>	<b>Pinobanksin<sup>a</sup></b>	8.67	271.06067	271.06120	1.96	253, 243, 165, 151, 107
<b>48</b>	<b>Isorhamnetin</b>	8.73	315.05057	315.05103	1.46	300, 151, 107
<b>49</b>	<b>Hesperetin<sup>a</sup></b>	8.77	301.07101	301.07176	2.49	271, 161
<b>50</b>	<b>Quercetin dimethyl ether 1</b>	8.98	329.06656	329.06668	0.36	315, 165
<b>51</b>	<b>Quercetin dimethyl ether 2</b>	9.64	329.06586	329.06668	2.49	315, 166
<b>52</b>	<b>Pinostrobin<sup>a</sup></b>	9.83	269.08121	269.08193	2.68	151, 179
<b>53</b>	<b>Prenyl caffeate</b>	9.85	247.09703	247.09758	2.23	135, 179
<b>54</b>	<b>Chrysin<sup>a</sup></b>	10.07	253.05009	253.05063	2.13	101, 151, 181, 209, 143
<b>55</b>	<b>Pinocembrin<sup>a</sup></b>	10.09	255.06563	255.06628	2.55	213, 211, 151
<b>56</b>	<b>Acacetin</b>	10.14	283.06094	283.06120	0.92	268, 133, 151, 107
<b>57</b>	<b>Caffeic acid phenethyl ester (CAPE)</b>	10.15	283.09714	283.09758	1.55	135, 179
<b>58</b>	<b>Galangin<sup>a</sup></b>	10.25	269.04489	269.04555	2.45	151, 183
<b>59</b>	<b>Genkwanin</b>	10.62	283.06042	283.06120	2.76	268, 239, 211

<b>60</b>	<b>Carnosol<sup>a</sup></b>	11.72	329.17487	329.17583	2.92	311, 296
<b>61</b>	<b>Carnosic acid<sup>a</sup></b>	12.81	331.19061	331.19148	2.63	287, 269

687 <sup>a</sup>Confirmed using available standards.

688 **Table 3** Quantification of individual polyphenolics (mg/kg), radical scavenging activity

689 (*RSA*) and total phenolic content (*TPC*) in the sage (*Salvia officinalis L.*) honeys.

	<b>SH1</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	<b>SH</b>	
		<b>2</b>	<b>SH3</b>	<b>4</b>	<b>SH</b>	<b>5</b>	<b>SH6</b>	<b>7</b>	<b>SH</b>	<b>8</b>	<b>9</b>	<b>SH</b>	<b>10</b>	<b>SH</b>	<b>11</b>	<b>SH</b>	<b>12</b>	<b>SH</b>
<b>Ga</b>	-	0.1		0.1	0.1		0.18	0.1	0.1	-	0.1		0.1		0.1	0.1	0.1	0.1
<b>A</b>		5		2	2			6	9	-	3		2		7	3	5	2
<b>GC</b>	-	0.4		0.1	0.1		0.16	-	-	0.1	0.2	0.1	0.1		-	-	0.2	0.2
		9		9	6					6	2	6	5				0	6
<b>Pr</b>	0.34	0.7		0.5	0.2		0.34	0.3	0.6	0.6	0.4	0.4	0.1	0.3	0.4	0.3	0.3	0.4
<b>A</b>		0		0.74	4	5		1	8	2	8	9	4	1	2	3	9	4
<b>EG</b>	0.12	-	0.46	0.1	0.0		0.12	0.0	0.1	0.1	0.1	0.0	0.0	0.1	0.0	0.0	-	0.2
<b>C</b>				0	8			8	0	4	4	9	7	6	8	8		4
<b>Ge</b>	-	-	-	-	-	-			0.1							0.0		0.0
<b>A</b>	-	-	-	-	-	-			3		-	-	-	-	-	-	1	-
<b>HB</b>	1.89	1.9		1.61	1.2	1.8		1.45	3.2	2.1	1.3	1.8	1.2	0.8	1.3	1.5	2.0	1.7
<b>A</b>		3			5	2			8	8	6	2	0	1	4	5	6	3
<b>Ch</b>	-	0.0		0.01	0.1	0.2			0.0	0.0	0.2	0.0	0.0	0.0	0.0	-	0.1	0.0
<b>A</b>		4			0	2			2	5	6	9	4	3	7	-	-	0
<b>C</b>	0.12	0.0		0.15	0.0	-			0.0	0.0	0.0	0.0	0.0	-	-	-	0.0	0.0
		3		9		-				5	1	4		5			1	6
<b>Ca</b>	0.48	1.8		0.56	0.3	0.4		0.59	0.5	0.9	0.5	0.6	0.6	0.6	0.3	0.3	0.4	0.7
<b>A</b>		9			7	8			7	2	4	4	1	2	7	8	7	0
<b>GC</b>	0.82	0.7		0.75	-	-			1.1	1.0	0.7	0.7	0.7	0.6	0.8	0.6	0.7	0.6
<b>G</b>		4			-	-			5	5	2	0	2	9	1	9	1	9
<b>EC</b>	0.10	-	0.03	0.0		6			-	-	-	-	-	-	-	-	0.0	0.0
																	5	7
<b>Co</b>	2.73	3.1		3.45	1.8	2.0		1.36	3.6	2.7	2.8	1.3	1.8	2.5	1.0	0.7	0.9	1.6
<b>A</b>		1			9	1			2	8	1	9	2	1	3	7	2	2
<b>Fe</b>	0.64	1.5		3.09	0.9	0.4		0.50	1.0	4.3	1.3	0.3	0.5	0.7	0.6	0.1	0.8	0.5
<b>A</b>		4			7	4			1	9	1	4	1	7	0	6	3	7
<b>Ro</b>	-	0.2		0.01	0.2	0.3		0.30	0.3	0.0	0.1	0.0	0.1	0.2	0.1	0.2	0.1	0.2
<b>A</b>		1			1	7			3	5	8	9	5	3	4	7	5	6
<b>EG</b>	0.94	1.2	0.97	0.8	0.7	1.11	1.0	0.9	0.8	-	0.9	1.2	0.9	0.5	1.0	1.2	1.1	1.6

<b>CG</b>	5	5	9	0	6	0	6	6	5	8	7	8	4	1
<b>Ab</b>	1.0	1.89	0.5	0.7	1.09	0.7	3.9	0.6	0.2	0.4	0.8	0.3	0.6	1.6
<b>A</b>	0.35	6	4	4	-	3	9	1	6	8	6	9	4	9
<b>RE</b>	-	0.1	-	0.2	-	-	-	-	-	-	-	-	0.0	0.4
<b>S</b>	-	1	-	2	-	-	-	-	-	-	-	-	8	6
<b>KA</b>	0.14	0.1	0.24	0.7	0.4	0.16	0.2	0.2	0.5	0.3	0.6	0.0	0.3	0.1
<b>E</b>	8	8	6	-	-	0	1	1	8	5	3	8	8	6
<b>PN</b>	-	2.3	0.35	0.3	0.6	1.57	1.0	1.8	0.8	1.3	1.0	2.2	0.4	0.5
<b>B</b>	-	5	3	3	-	0	2	2	0	0	6	4	8	0
<b>QU</b>	0.07	0.3	0.14	0.5	0.2	0.12	0.1	0.3	0.3	1.0	0.3	0.1	0.3	0.1
<b>E</b>	8	8	3	-	-	7	3	6	5	6	1	2	6	4
<b>CH</b>	0.06	1.9	0.41	0.2	0.2	0.87	0.4	0.9	0.4	0.8	0.7	1.5	0.4	0.9
<b>R</b>	8	7	3	-	-	8	5	7	1	3	0	2	7	0
<b>PN</b>	-	0.3	-	-	-	0.07	0.0	0.1	0.0	0.0	0.0	0.1	-	0.0
<b>S</b>	-	4	-	-	-	-	4	9	1	7	3	9	-	5
<b>PN</b>	-	0.8	0.09	0.0	0.1	0.51	0.2	0.4	0.2	0.4	0.3	0.7	0.1	0.1
<b>C</b>	-	0	5	5	-	2	6	8	5	5	2	5	7	6
<b>HE</b>	-	0.8	0.19	0.0	0.2	0.37	0.1	0.3	0.2	0.3	0.2	0.5	0.0	0.0
<b>S</b>	-	4	6	0	-	1	8	7	3	0	2	4	9	6
<b>GL</b>	-	0.3	0.01	-	0.0	0.11	0.0	0.2	0.0	0.1	0.0	0.2	0.0	0.0
<b>N</b>	-	9	-	-	1	-	7	3	4	2	7	2	1	-
<b>TP</b>	553.	485	424.	591	522	484.	471	509	538	56	747	417	525	444
<b>C</b>	98	.86	.92	.08	.33	91	.03	.11	.51	0.7	.49	.73	.39	.97
										2				.54
<b>RS</b>	819.	627	526.	770	548	571.	541	627	610	58	894	474	641	675
<b>A</b>	75	.44	81	.65	.55	41	.69	.79	.38	6.0	.87	.17	.38	.23
										4				.81
														.20
														.11
														.20

690 **GaA** – Gallic acid; **GC** – Gallocatechin; **PrA** – Protocatechuic acid; **EGC** – Epigallocatechin; **GeA** – Gentisic  
691 acid; **HBA** – *p*-Hydroxybenzoic acid; **ChA** – Chlorogenic acid; **C** – Catechin; **CaA** – Caffeic acid; **GGC** -  
692 Gallocatechin gallate; **EC** – Epicatechin; **CoA** – *p*-Coumaric acid; **FeA** – Ferulic acid; **RoA** – Rosmarinic acid;  
693 **EGCG** - Epigallocatechin gallate; **AbA** – *cis*, *trans*-Abscisic acid; **RES** – Resveratrol; **KAE** – Kaempferol;  
694 **PNB** – Pinobanksin; **QUE** – Quercetin; **CHR** – Chrysins; **PNS** – Pinostrobin; **PNC** – Pinocembrin; **HES** –  
695 Hesperetin; **GLN** – Galangin; **TPC** – Total phenolic content (mg GEA/kg); **RSA** – Radical scavenging activity  
696 ( $\mu\text{mol TE/kg}$ ).

**Table 4** Quantification of the sugars and sugar alcohols in the sage (*Salvia officinalis* L.) honeys (g/kg).

	<b>SH1</b>	<b>SH2</b>	<b>SH3</b>	<b>SH4</b>	<b>SH5</b>	<b>SH6</b>	<b>SH7</b>	<b>SH8</b>	<b>SH9</b>	<b>SH10</b>	<b>SH11</b>	<b>SH12</b>	<b>SH13</b>	<b>SH14</b>	<b>SH15</b>	<b>SH16</b>	<b>SH17</b>	<b>SH18</b>
<b>ERY</b>	0.04	0.07	0.05	0.05	0.02	0.07	0.06	0.07	0.05	0.07	0.09	0.69	0.06	0.07	0.18	0.12	0.11	0.09
<b>SOR</b>	0.04	0.13	0.03	0.06	0.09	0.08	0.30	0.33	0.20	0.06	0.06	0.03	0.04	0.02	0.18	0.05	0.05	0.02
<b>TRE</b>	1.23	5.70	2.71	2.07	5.34	0.21	5.07	2.76	1.45	5.08	1.82	0.75	1.31	2.39	1.41	0.14	0.63	1.02
<b>ARA</b>	0.04	0.15	0.05	0.10	0.10	0.37	0.12	0.07	0.06	0.07	0.10	0.05	0.09	0.05	0.05	0.06	0.08	0.03
<b>GLU</b>	305.03	272.07	280.3	253.24	212.95	206.81	108.89	245.39	200.35	263.82	227.38	240.15	244.46	252.72	271.21	262.22	277.59	263.08
<b>FRU</b>	399.41	464.01	440.3	420.83	393.63	475.99	480.76	437.51	442.23	462.32	464.06	489.06	461.97	455.68	461.76	464.87	447.35	471.03
<b>SUC</b>	30.41	28.78	11.02	15.69	14.17	14.27	20.71	14.03	11.87	16.54	21.4	27.08	25.64	19.01	11.21	18.59	16.34	16.16
<b>TUR</b>	1.34	0.12	0.32	0.68	0.6	0.83	1.13	0.76	0.88	0.65	0.87	0.82	0.82	0.90	0.11	0.77	1.16	1.06
<b>GLY</b>	1.51	0.11	0.16	0.09	0.12	0.17	0.12	0.16	0.03	0.19	0.10	0.05	0.08	0.07	0.17	0.12	0.12	0.13
<b>GAL</b>	0.06	0.06	0.04	0.09	0.46	0.33	0.20	0.36	0.02	0.07	0.08	0.03	0.02	0.03	0.2	0.06	0.07	0.03
<b>iMAL</b>	4.45	3.11	5.31	14.6	3.63	7.13	7.68	5.40	3.95	12.61	12.34	3.52	7.44	8.51	8.28	10.22	11.62	9.98
<b>iMALt</b>	1.44	2.88	0.73	4.66	2.55	2.76	1.24	1.48	1.11	3.79	5.66	1.11	0.33	0.36	0.31	2.41	3.91	2.44
<b>MAL</b>	6.05	3.74	5.92	16.85	8.76	9.32	11.91	7.32	7.38	11.77	11.09	7.40	8.98	10.06	8.33	11.49	12.31	9.91
<b>MALT</b>	0.11	0.06	0.01	0.02	0.02	0.05	0.06	0.08	0.06	0.04	0.07	0.08	0.09	0.07	0.02	0.09	0.08	0.08
<b>SUM</b>	751.16	780.99	746.95	729.03	642.44	718.39	638.25	715.72	669.64	777.08	745.12	770.82	751.33	749.94	763.42	771.21	771.42	775.06
<b>FRU/GLU</b>	1.31	1.71	1.57	1.66	1.85	2.30	4.42	1.78	2.21	1.75	2.04	2.04	1.89	1.80	1.70	1.77	1.61	1.79

699	<i>MAL/iMAL</i>	1.36	1.20	1.11	1.15	2.41	1.31	1.55	1.35	1.87	0.93	0.90	2.10	1.21	1.18	1.01	1.13	1.06	0.99
700	<i>ERY</i> – Erythritol; <i>SOR</i> – Sorbitol; <i>TRE</i> – Trehalose; <i>ARA</i> – Arabinose; <i>GLU</i> – Glucose; <i>FRU</i> – Fructose; <i>SUC</i> – Sucrose; <i>TUR</i> – Turanose; <i>GLY</i> – Glycerol; <i>GAL</i> – Galactitol; <i>iMAL</i> – Isomaltose; <i>iMALT</i> – Isomaltotriose; <b>MAL</b> – Maltose; <b>MALT</b> – Maltotriose; <b>SUM</b> – Summary of quantified sugars and sugar alcohols; <i>FRU/GLU</i> – Fructose/Glucose ratio; <i>MAL/iMAL</i> – Maltose/Isomaltose ratio.																		
701																			

**Table 5** Quantification of the minerals in the sage (*Salvia officinalis L.*) honeys (mg/kg).

	SH 1	SH 2	SH 3	SH 4	SH 5	SH 6	SH 7	SH 8	SH 9	SH 10	SH 11	SH 12	SH 13	SH 14	SH 15	SH 16	SH 17	SH 18
<b>A</b>	0.0	0.4	0.4	0.4	0.1	0.0	<L	0.2	0.5	0.2	0.3	<L	0.3	0.8	0.0	0.2	0.1	0.1
<b>I</b>	95	33	31	19	45	23	OQ	61	05	66	87	OQ	06	83	59	63	49	11
<b>A</b>	0.0	<L	<L	<L	<L	<L	0.0	<L	<L	<L	<L	<L	<L	<L	0.0	<L	<L	
<b>s</b>	03	OQ	OQ	OQ	OQ	OQ	06	OQ	OQ	OQ	OQ	OQ	OQ	OQ	13	OQ	OQ	
<b>B</b>	1.2	2.3	1.1	1.2	1.8	2.7	2.1	2.5	1.9	1.3	2.0	2.2	2.1	1.7	3.0	2.1	1.4	1.6
	70	76	34	90	76	60	13	99	07	88	86	01	74	74	29	40	55	81
<b>B</b>	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
<b>a</b>	51	94	84	68	76	66	59	76	67	64	72	76	64	71	71	60	69	62
<b>C</b>	23.	49.	50.	42	50.	36.	23.	31.	47.	34.	32.	39.	20.	35.	22.	36.	30.	20.
<b>a</b>	823	894	471	7	787	591	557	122	375	747	980	409	098	978	471	205	493	885
<b>C</b>	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
<b>d</b>	03	04	03	04	03	03	03	04	03	03	03	03	03	04	04	03	03	03
<b>C</b>	<L	<L	<L	<L	<L	<L	<L	<L	<L									
<b>o</b>	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ									
<b>C</b>	0.0	0.0	0.0	<L	<L	<L	<L	0.0	0.0	0.0	0.0	<L	0.0	0.0	<L	<L	<L	0.0
<b>r</b>	04	05	04	OQ	OQ	OQ	OQ	15	06	10	07	OQ	13	09	OQ	OQ	OQ	03
<b>C</b>	0.1	0.2	0.1	0.1	0.2	0.1	0.2	0.1	0.2	0.1	0.2	0.1	0.1	0.3	0.1	0.1	0.1	0.1
<b>u</b>	01	99	87	26	20	98	83	73	33	55	19	21	66	87	13	52	24	09
<b>F</b>	0.2	0.9	0.5	0.5	0.4	0.4	0.2	0.8	0.3	0.4	2.2	0.4	2.4	0.8	0.1	0.3	0.2	0.4
<b>e</b>	47	28	27	33	57	42	74	53	71	79	78	00	94	06	95	82	48	96
<b>K</b>	2.1	1.2	1.8	2.0	1.3	0.8	0.9	0.9	1.0	1.1	1.8	0.5	1.3	0.7	1.2	1.1	2.1	0.6
<b>“</b>	51	81	38	37	13	44	09	82	26	16	73	92	00	30	13	45	07	75
<b>L</b>	<L	<L	<L	<L	<L	<L	<L	<L	<L									
<b>i</b>	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ									
<b>M</b>	5.0	26.	11.	10.	7.9	7.9	8.9	25.	8.0	11.	8.7	5.9	7.1	9.9	10.	7.1	5.7	10.